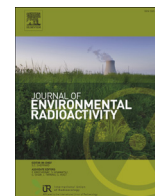


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## Distribution and risk assessment of radionuclides released by Fukushima nuclear accident at the northwest Pacific

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## ABSTRACT

In order to understand the impact of Fukushima Nuclear Accident (FNA) on the marine environment, seawater and a composite squid (*Ommastrephes bartrami*) sample were collected on the monitoring cruise XT01 during June 16–July 4, 2011. The concentration levels of Cesium-134, Cesium-137, Strontium-90, Silver-110m, Cobalt-58 and Cobalt-60 were measured both for the seawater and squid samples. The elevated activity levels of Cesium-134 and Cesium-137 were found in the sampling area. Cesium-134 and Silver-110m, which were usually undetectable before FNA, were also found in the squid sample, with the activity levels of  $1.65 \pm 0.13$  Bq/kg-wet and  $0.07 \pm 0.01$  Bq/kg-wet, respectively. The radiological assessment result showed that the radioactive release from the FNA would not have a significant adverse effect on marine biota at the population level.

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## 1. Introduction

On March 11 05:46 UTC, 2011, a 9.0 magnitude ( $M_W$ ) earthquake occurred at northwest Pacific (epicenter at 38.1°N and 142.9°E, 130 km ESE of the Ojika Peninsula of Japan). The subsequent tsunami triggered the accident at the Fukushima nuclear power plant (FNPP), which was the first major accident to directly discharge huge liquid radioactive contaminants into the marine environment. The amount of radioactive contaminants released directly into the marine environment was estimated to be 3–27 PBq for  $^{137}\text{Cs}$  (Bailly du Bois et al., 2012; Kawamura et al., 2011; Masumoto et al., 2012; Rypina et al., 2013; Tsumune et al., 2012), 11 PBq for  $^{131}\text{I}$  (Kawamura et al., 2011), 0.1 PBq for  $^3\text{H}$  (Povinec et al., 2013), 2.35–7 GBq for  $^{129}\text{I}$  (Hou et al., 2013; Povinec et al., 2013) and 0.08–0.9 PBq for  $^{90}\text{Sr}$  (Casacuberta et al., 2013; Periañez et al., 2013).

Although ocean has a great capacity to dilute and disperse the radioactive release due to its large volume, the long half-life radionuclides will stay in the marine environment for a long period and possibly threaten the marine ecosystem or human-being via food chain transferring, especially for the coastal benthic environment (Wada et al., 2013).

After the discharge of the radioactive contaminants, a sophisticated physical–biogeochemical model should be set up to give a comprehensive evaluation of radioactivity on marine ecosystem (Maderich et al., 2014). Parameters in the model should be determined by field sampling. The data from field surveys could also be applied to validate the model. However, the scarcity of field data in different environmental matrices would constrain the prediction ability of the model, especially in the open ocean.

In order to understand the fate of the radioactive contaminants after the Fukushima nuclear accident (FNA) and to assess the relevant effect and radiological risk on the open ocean in the northwest Pacific, the XT01 cruise was implemented by the Third Institute of Oceanography, State Oceanic Administration of China (SOAC) during June 16–July 4, 2011. The monitoring region was at 145.007–149.117°E and 34.015–39.997°N. Surface seawater samples were collected at 35 stations (forming 4 sections) for radioactivity measurement in the cruise. Meanwhile the biota sample was collected at 1 station (33.492°N, 148.006°E).

## 2. Methods and materials

## 2.1. Sampling sites

The two main current systems in this oceanic region are the Kuroshio Current extension with high temperature and high salinity and the Oyashio Current with low temperature and low

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salinity (Shimizu et al., 2001). The Oyashio Current, flowing southwards along the east coast of Japan, joins with the northward Kuroshio Current at 35°N and flows eastwards into the open ocean. The historical data of spatial–temporal hydrodynamics of the fronts of temperature and salinity were obtained by remote sensing at the monitoring region to determine the monitoring region and sampling sites before the cruise. The monitoring region was located to the east of Japanese EEZ, mainly at the northern side of the Kuroshio extension, which is on the main pathway of the FNA radioactive release transportation. 35 sampling sites were set, forming 3 latitudinal sections and 1 longitudinal section.

The monitoring region's average surface dynamic topography during June–July of 2011, the accordingly calculated geostrophic flows and the sampling stations of seawater and biota are shown in Fig. 1, where the color represents the surface dynamic topography (in centimeters), the arrows represent the geostrophic flow (the ones with velocities lower than 10 cm/s were not showed), the dots represent the seawater sampling stations and the triangle represents the biota sampling station. Detailed information of the sampling stations is given in Table 1.

The surface dynamic topography data were the quasi-real-time multi-source integrated altimetry data from T/P, Jason and ERS1/2 satellites provided by AVISO ([www.aviso.oceanobs.com/](http://www.aviso.oceanobs.com/)). The spatial resolution of the data was 1/3°, and the time resolution was 7 d, with the correction for tides and sea level pressure. The dynamic topography was established according the method proposed by Li et al. (2002) using satellite altimetry data combined with climatological temperature and salinity data (WOA01).

At the main axis of the Kuroshio extension, the flow velocity reached over 100 cm/s, while the flow velocity was much lower at the cyclonic and anti-cyclonic mesoscale eddies around the Kuroshio extension.

## 2.2. Sampling methods

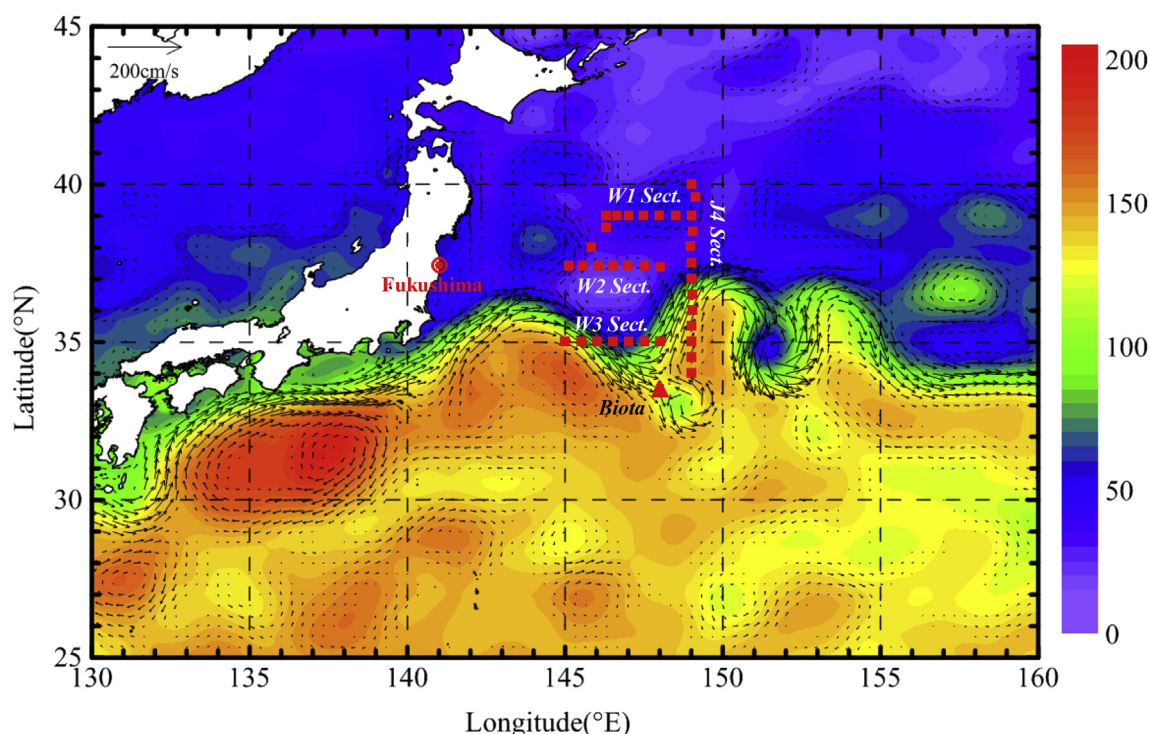
Surface seawater was sampled by submersible pumps at the depths of 0, 20 and 50 m. For each, 150 L seawater samples were collected, stored in polyethylene barrels with acidification to pH = 2, and taken back to land-base laboratory for analysis.

Net tows were applied for several times at different locations but failed to obtain enough sample for radionuclides analysis, so angling with hooks was used for biota sampling and succeeded to get 11.55 kg of squid (*Ommastrephes bartramii*) as a single composite sample.

## 2.3. Analysis and detection methods

### 2.3.1. Gamma-emitting radionuclides ( $^{110m}\text{Ag}$ , $^{134,137}\text{Cs}$ , $^{58,60}\text{Co}$ ) in seawater

Aliquots of 60 L clear seawater were placed into open polypropylene drums.  $\text{Ag}^+$  carrier (1.2 g,  $\text{AgNO}_3$ ) was added and stirred for 30 min to make a  $\text{AgCl}$  precipitation, then left for 3 days settling. The clear supernatant was then siphoned into another bucket; 30 mg  $\text{Cs}^+$  carrier ( $\text{CsCl}$ ) was added and adjusted to pH < 2; next, 15 g AMP (Ammonium Molybdophosphate) was added and stirred for 30 min to make the cesium precipitate, allowing 2 days for settling. The clear supernatant was siphoned into a third bucket; 100 mg  $\text{Co}^{2+}$  carrier and 200 mg  $\text{Fe}^{3+}$  were added and stirred. Then  $\text{NaOH}$  was added until the solution turned pink (pH ~ 8) and stirred for 30 min to make the  $\text{Fe}(\text{OH})_3$  co-precipitation. The lower precipitation phase of the three buckets was suction filtered using quantitative filter paper; the papers were transferred into crucibles and placed in a muffle furnace at 450 °C for 2 h; the ashes were weighed, porphyzied and boxed, then subjected to HPGe spectrometry.



**Fig. 1.** Map of surface dynamic topography, geostrophic flows and the sampling stations (color – surface dynamic topography (cm); arrows – geostrophic flows; squares – seawater sampling stations; and triangle – biota sampling station).

**Table 1**  
Information of the sampling stations.

| No. | Section | Station | Sampling date | Longitude (E) | Latitude (N) |
|-----|---------|---------|---------------|---------------|--------------|
| 1   | W3      | W3-3    | 06/21/2011    | 145.007°      | 34.992°      |
| 2   |         | W3-4    | 06/21/2011    | 145.505°      | 34.990°      |
| 3   |         | W3-5    | 06/21/2011    | 146.002°      | 34.997°      |
| 4   |         | W3-6    | 06/21/2011    | 146.498°      | 35.000°      |
| 5   |         | W3-7    | 06/21/2011    | 146.993°      | 35.003°      |
| 6   |         | W3-8    | 06/22/2011    | 147.487°      | 35.003°      |
| 7   |         | W3-9    | 06/22/2011    | 147.970°      | 35.008°      |
| 8   | W2      | W2-7    | 06/22/2011    | 147.997°      | 37.380°      |
| 9   |         | W2-6    | 06/22/2011    | 147.505°      | 37.405°      |
| 10  |         | W2-5    | 06/22/2011    | 147.005°      | 37.400°      |
| 11  |         | W2-4    | 06/22/2011    | 146.507°      | 37.398°      |
| 12  |         | W2-3    | 06/23/2011    | 146.032°      | 37.408°      |
| 13  |         | W2-2    | 06/23/2011    | 145.543°      | 37.398°      |
| 14  |         | W2-1    | 06/23/2011    | 145.098°      | 37.397°      |
| 15  | W1      | Wa      | 06/23/2011    | 145.822°      | 37.987°      |
| 16  |         | Wb      | 06/23/2011    | 146.272°      | 38.618°      |
| 17  |         | W1-1    | 06/23/2011    | 146.327°      | 38.998°      |
| 18  |         | W1-2    | 06/23/2011    | 146.660°      | 39.008°      |
| 19  |         | W1-3    | 06/23/2011    | 146.993°      | 39.007°      |
| 20  |         | W1-4    | 06/24/2011    | 147.467°      | 39.000°      |
| 21  |         | W1-5    | 06/24/2011    | 147.975°      | 39.010°      |
| 22  |         | W1-6    | 06/24/2011    | 148.495°      | 39.002°      |
| 23  | J4      | J4-13   | 06/24/2011    | 149.005°      | 39.997°      |
| 24  |         | J4-12   | 06/24/2011    | 149.117°      | 39.540°      |
| 25  |         | J4-11   | 06/24/2011    | 149.008°      | 38.995°      |
| 26  |         | J4-10   | 06/24/2011    | 149.023°      | 38.498°      |
| 27  |         | J4-9    | 06/25/2011    | 148.955°      | 38.032°      |
| 28  |         | J4-8    | 06/25/2011    | 149.000°      | 37.502°      |
| 29  |         | J4-7    | 06/25/2011    | 149.017°      | 37.003°      |
| 30  |         | J4-6    | 06/25/2011    | 149.015°      | 36.510°      |
| 31  |         | J4-5    | 06/26/2011    | 149.012°      | 36.010°      |
| 32  |         | J4-4    | 06/26/2011    | 149.008°      | 35.497°      |
| 33  |         | J4-3    | 06/26/2011    | 149.003°      | 34.993°      |
| 34  |         | J4-2    | 06/26/2011    | 149.003°      | 34.505°      |
| 35  |         | J4-1    | 06/26/2011    | 148.997°      | 34.015°      |
| 36  | Biota   | B01     | 06/26/2011    | 148.006°      | 33.492°      |

### 2.3.2. Beta-emitting radionuclide $^{90}\text{Sr}/^{90}\text{Y}$ in seawater

Aliquots of 40 L clear seawater were added with 200 mg  $\text{Sr}^{2+}$  carrier, 40 mg  $\text{Y}^{3+}$  carrier, 200 g anhydrous sodium carbonate and 30 g solid ammonium chloride while stirring, then left for 2 days settling. The supernatant was removed and the precipitate was collected with suction filtration. The precipitate was then dissolved with 6 M  $\text{HNO}_3$  and adjusted to pH = 1.  $^{90}\text{Sr}$  and its daughter  $^{90}\text{Y}$  was separated by two extractions with bis(2-ethylhexyl) phosphoric acid (HDEHP)—normal heptane (1:9) 50 mL under pH ~ 1.0;  $^{90}\text{Y}$  was back extracted from the organic phase with 9 mol/L HCl, precipitated with aqua ammonia, and dissolved in  $\text{HNO}_3$ . 5 mL saturated oxalic acid was added into the solution and pH was adjusted to 1.5–2.0 with  $\text{NH}_3 \cdot \text{H}_2\text{O}$  to make  $\text{Y}_2(\text{C}_2\text{O}_4)_3 \cdot 9\text{H}_2\text{O}$  precipitation. The precipitate was collected with suction filtration, dried in oven at 50 °C, weighed and then subject to low-level  $\alpha/\beta$  counter.

### 2.3.3. $^{131}\text{I}$ in seawater

Aliquots of 10 L seawater were added with 20 mg KI carrier. All Iodine was reduced to  $\text{I}^-$  with  $\text{NaHSO}_3$  and  $\text{HNO}_3$ . The  $\text{I}^-$  was concentrated with strongly basic anion exchange resin ( $201 \times 7 \text{ NO}_3^-$ ) column and the column was washed with  $\text{H}_2\text{O}$  and 3 mol/L  $\text{NaNO}_3$ . The  $\text{I}^-$  was washed down with 0.5 mol/L  $\text{NaNO}_3$ .  $\text{NaNO}_2$  was added into the solution and  $\text{I}_2$  was then extracted with  $\text{CCl}_4$ ;  $\text{NaHSO}_3$  was added into extract liquor and  $\text{I}^-$  was back-extracted with  $\text{H}_2\text{O}$ ;  $\text{AgNO}_3$  was added to cause  $\text{AgI}$  precipitation. The precipitate was collected with suction filtration, dried in oven at 50 °C, weighed and then subject to low-level  $\alpha/\beta$  counter.

### 2.3.4. Gamma-emitting radionuclides ( $^{110m}\text{Ag}$ , $^{134,137}\text{Cs}$ , $^{58,60}\text{Co}$ , etc) in squid

All the squid were analyzed as one sample. The whole bodies were weighed fresh, dried at 50 °C, and then ashed at 450 °C. The ash was porphyzied, weighed and boxed, then subjected to HPGe spectrometry.

### 2.3.5. $^{90}\text{Sr}/^{90}\text{Y}$ in squid

An aliquot of 10 g squid ash was added with 200 mg  $\text{Sr}^{2+}$  carrier, 40 mg  $\text{Y}^{3+}$  carrier, 50 mL concentrated nitric acid and 5 mL concentrated  $\text{H}_2\text{O}_2$ . After boiling for 30 min, the sample was filtered and saturated  $\text{Na}_2\text{CO}_3$  was added into the supernatant. The precipitate was collected by suction filtration, dissolved with  $\text{HNO}_3$  and then adjusted pH to 1. The following procedures were the same as the analyzing method for  $^{90}\text{Sr}/^{90}\text{Y}$  in seawater.

## 3. Results and discussions

### 3.1. Background activity of $^{137}\text{Cs}$ , $^{134}\text{Cs}$ and $^{90}\text{Sr}$ at the northwest Pacific

$^{137}\text{Cs}$  and  $^{134}\text{Cs}$  are two of the most important radioactive contaminants released in a nuclear power plant accident due to their large fission yield and relatively long half-life (30.2 y and 2.1 y respectively). The atomic bomb testing and Chernobyl nuclear power plant accident in the last century have been the largest sources for the  $^{137}\text{Cs}$  in the marine environment (IAEA, 2004). According to the data from IAEA's MARIS (Marine Information System) database, from the year 2000–2010, the radioactivity of  $^{137}\text{Cs}$  in surface seawater near Japan ranged from 0.04 to 3.4 mBq/L, on average  $1.7 \pm 0.6$  mBq/L ( $n = 961$ ) in this region.  $^{134}\text{Cs}$  is usually not detectable in the surface seawater because there was no new major source in the past 20 years and the  $^{134}\text{Cs}$  released in last century has decayed to extremely low levels, well below the radioanalytical detection limit. Therefore,  $^{134}\text{Cs}$  could be considered as an indicator of contaminants released from the recent FNA.

$^{90}\text{Sr}$  is another one of the important radionuclides released in an accident because of its relatively long half-life (28.6 y) and high affinity to the bones of biota. The earlier atomic bomb testing and Chernobyl accident also released large amounts of  $^{90}\text{Sr}$  into the marine environment. The pre-Fukushima background activity of  $^{90}\text{Sr}$  in surface seawater at the northwest Pacific is 0.01–2.6 mBq/L, on average  $1.2 \pm 0.4$  ( $n = 871$ ) (from IAEA's MARIS Database).

### 3.2. Distribution of $^{137}\text{Cs}$ and $^{134}\text{Cs}$ in seawater

The results showed that the radioactivity of  $^{137}\text{Cs}$  in surface seawater varied widely, ranging from 1 to 826 mBq/L, of which 65% data was lower than 100 mBq/L, while only 1% was higher than 800 mBq/L.  $^{134}\text{Cs}$  activity data were distributed in a similar pattern (Fig. 2). The median value of  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$  activity in seawater was 47.1 mBq/L and 39.5 mBq/L, respectively. At the 0 m depth layer, the  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$  activity ranged from 1 to 826 mBq/L and from below detection limit to 757 mBq/L, and the medians were 51 mBq/L and 45 mBq/L, respectively. At the 20 m depth layer, the activity of  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$  ranged from 1 to 755 mBq/L and from below detection limit to 662 mBq/L, and the medians were 39 mBq/L and 35 mBq/L, respectively. At the depth of 50 m, the activity of  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$  varied from 2 to 678 mBq/L and from below detection limit to 597 mBq/L, and the medians were 52 mBq/L and 43 mBq/L, respectively.

In Fig. 3(a)–(f), the horizontal distributions of  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$  at different depths are shown. The distribution patterns for  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$  were extremely similar due to their same biogeochemical behavior. Both of the  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$  activities were higher in the



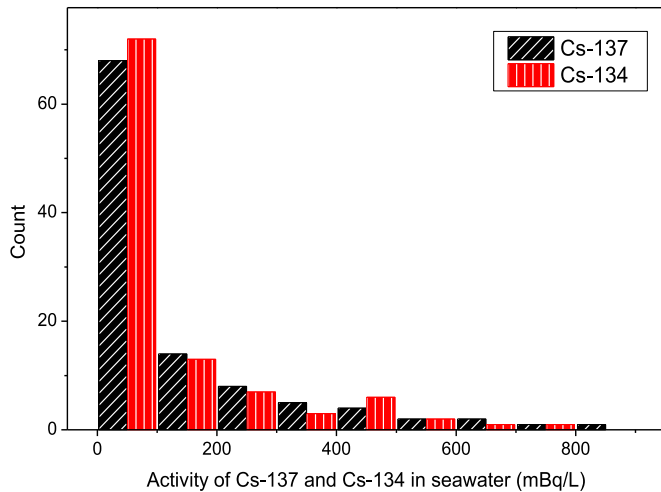


Fig. 2. Histogram of  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$  activity in seawater.

surface seawater at the middle and east of section W2 than in the other regions. The largest value was observed at the station W2-7, over 615 km (332 NM) away from Fukushima. The distribution patterns were similar in the depths of 20 m and 50 m as in the surface seawater.

The profile of  $^{137}\text{Cs}$  activity in seawater at Section J4 (Fig. 4) showed that the seawater  $^{137}\text{Cs}$  activity was higher at the northern

side of the Kuroshio extension ( $35^\circ\text{N}$ ) than at the southern side, revealing the Kuroshio extension's barrier effect on FNA radionuclide transport. However, part of the radionuclide was transported southwards across the Kuroshio extension with the north Pacific mode water (Men et al., 2015).

The  $^{134}\text{Cs}$  activity was decay-corrected to Mar 11 2011 and plotted against  $^{137}\text{Cs}$  activity in Fig. 5. The slope of the linear fit was 0.983, very close to the  $^{134}\text{Cs}/^{137}\text{Cs}$  ratio (1, reported by Masson et al. (2011), Buesseler et al. (2012) and Merz et al. (2013)) of FNA, revealing that the radionuclides were released from FNA.

Compared with the averaged background data, the highest activity of  $^{137}\text{Cs}$  observed in surface seawater was 485 times above background, and the observed median activity of  $^{137}\text{Cs}$  was 28 times higher than the background. Also, similar levels of  $^{134}\text{Cs}$  were detected at the monitoring region, which are clearly the contamination released by Fukushima accident.

### 3.3. Distribution of $^{90}\text{Sr}$ in the seawater

Due to the complicated and laborious analytical method for  $^{90}\text{Sr}$ , the radioactivity of  $^{90}\text{Sr}$  in seawater was less reported and evaluated after the FNA (Casacuberta et al., 2013; Povinec et al., 2012). The seawater  $^{90}\text{Sr}$  activity in the monitoring region ranged from 1 to 31 mBq/L, and the median was 2 mBq/L. At the depth of 0 m, 20 m and 50 m, all the minimum activities of  $^{90}\text{Sr}$  were 1 mBq/L. The maximum activity of  $^{90}\text{Sr}$  at these depths was 31 mBq/L, 26 mBq/L and 29 mBq/L, and the medians were 2 mBq/L, 2 mBq/L and 3 mBq/L, respectively.

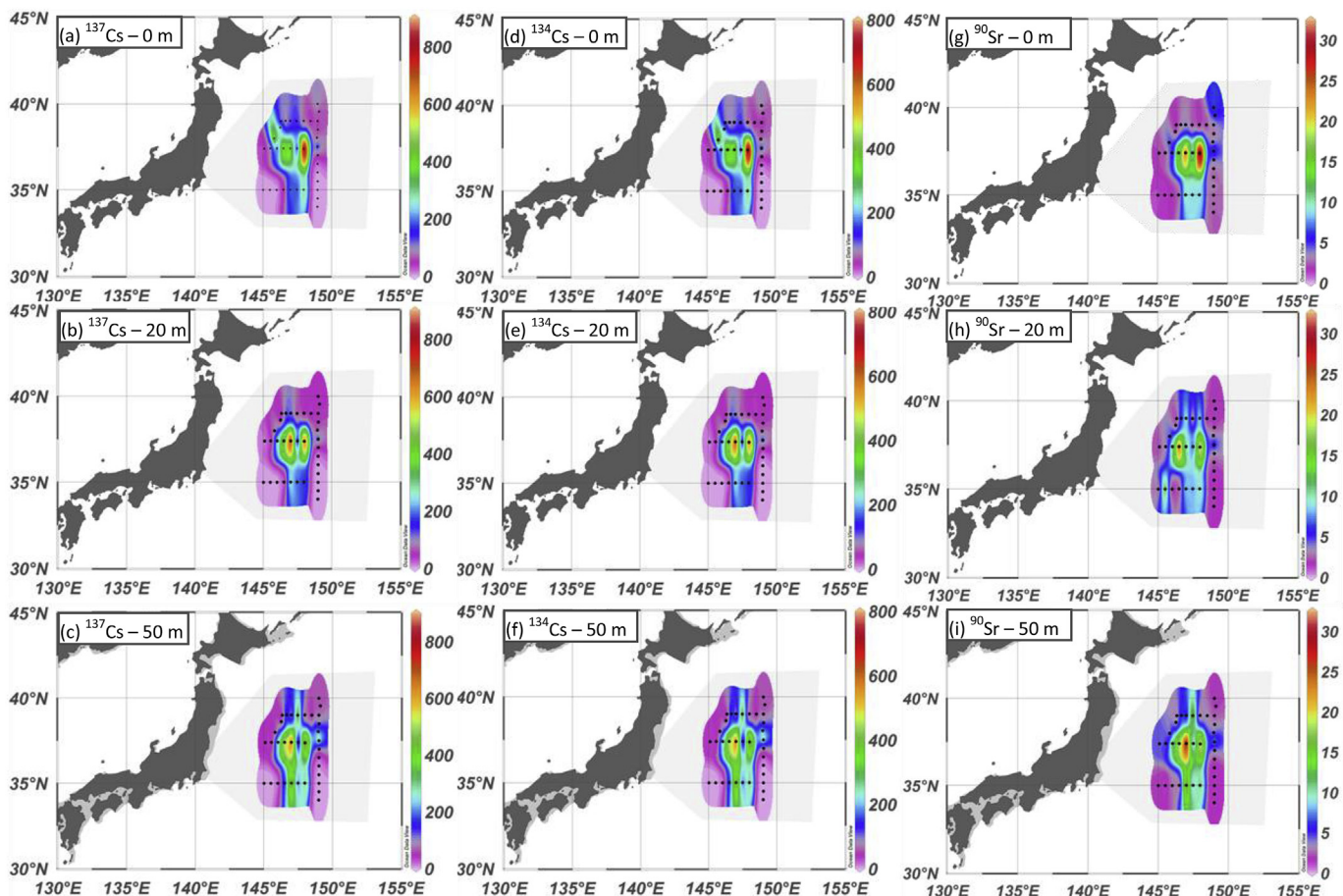


Fig. 3. Horizontal distributions of  $^{137}\text{Cs}$ ,  $^{134}\text{Cs}$  and  $^{90}\text{Sr}$  (mBq/L) in seawater.

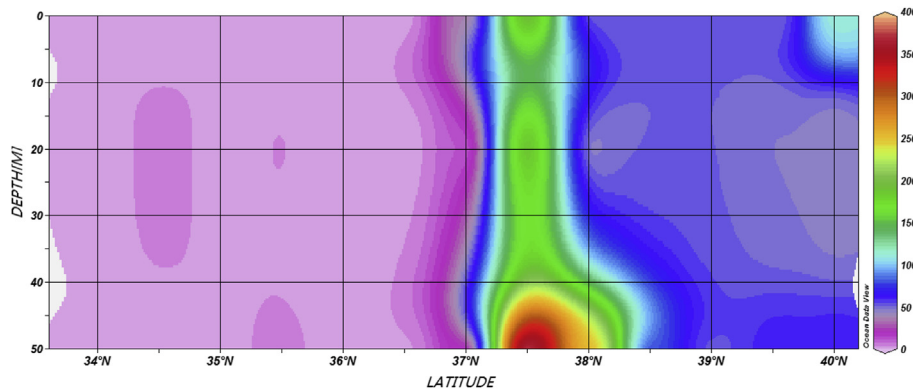


Fig. 4. Profile of  $^{137}\text{Cs}$  activity (mBq/L) in seawater at Section J4.

The horizontal distributions of  $^{90}\text{Sr}$  at different depths are shown in Fig. 3(g)–(i). The distribution pattern of  $^{90}\text{Sr}$  was similar to those of  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$ . The activities of  $^{90}\text{Sr}$  were also higher at the middle and east part of section W2, with the highest value observed at station W2-7.

The measured  $^{90}\text{Sr}$  activity was plotted against  $^{137}\text{Cs}$  activity in Fig. 5. The slope of the linear fit was 0.029, comparable with the  $^{90}\text{Sr}/^{137}\text{Cs}$  ratio (0.01–0.02) reported by Buesseler et al. (2013) and Periañez et al. (2013). The  $R^2$  of the linear fit was 0.727, lower than the  $^{134}\text{Cs}/^{137}\text{Cs}$  linear fit  $R^2$ , probably because the difference between the biogeochemical behavior of Sr and Cs, as well as the different time distribution of the Sr and Cs releases.

Compared with the background data mentioned in Section 3.1, the observed highest  $^{90}\text{Sr}$  activity (31 mBq/L) was as much as 26 times the average background and the observed median  $^{90}\text{Sr}$  activity was twice of the average background activity.

### 3.4. Other radionuclides in the seawater

Other radionuclides, such as  $^{110\text{m}}\text{Ag}$ ,  $^{58}\text{Co}$  and  $^{60}\text{Co}$  were analyzed but the results were below detection limits (0.36 mBq/L for  $^{110\text{m}}\text{Ag}$ , 0.36 mBq/L for  $^{58}\text{Co}$  and 0.41 mBq/L for  $^{60}\text{Co}$ , respectively).

### 3.5. Activities of radionuclides in squid

The biota sample obtained in the cruise was of squid (*O. bartrami*). The body length of the samples varied from 161 to 468 mm, on average 251 mm, and the weight varied from 94 to 3102 g, on

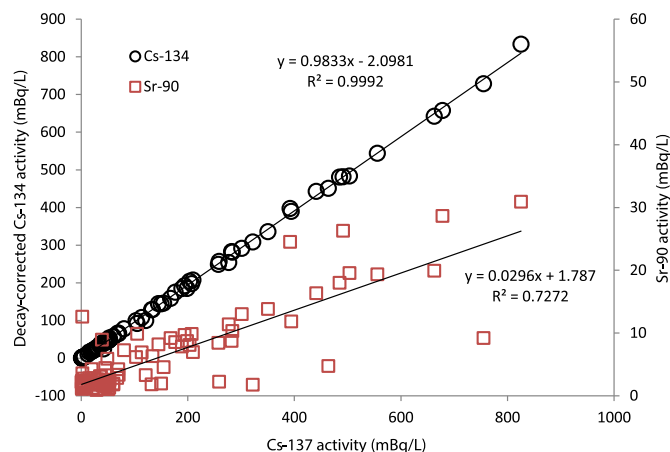


Fig. 5. Decay-corrected  $^{134}\text{Cs}$  and  $^{90}\text{Sr}$  activity plotted against  $^{137}\text{Cs}$  activity.

average 594 g. For 61% of the samples, the body length varied from 160 to 230 mm, 39% longer than 310 mm, according to which it could be inferred that the age of the samples varied from 3 to 7 months. The wet weight of the composite sample was 11.55 kg, and the weights were 2.75 kg and 19.13 g respectively after drying and ashing.

The activity of  $^{90}\text{Sr}$  in the squid sample was  $7.64 \pm 1.67$  Bq/kg-wet, the activity of  $^{110\text{m}}\text{Ag}$  was  $1.65 \pm 0.13$  Bq/kg-wet, and the activity of  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$  was  $0.06 \pm 0.01$  Bq/kg-wet and  $0.07 \pm 0.01$  Bq/kg-wet, respectively.

$^{110\text{m}}\text{Ag}$  is one of the main activation products in the effluent of nuclear power stations, and were reported after the FNA (Lepage et al., 2014; Saegusa et al., 2013). It has a relatively short half-life of 250 d, so usually it's not detectable in the normal marine biota. It's probable that the  $^{110\text{m}}\text{Ag}$  in the squid sample came from the contaminants released in the FNA because cephalopods have a long migratory path and potentially high concentration factor (CF) for  $^{110\text{m}}\text{Ag}$ , similar to zooplankton. Although IAEA (2004) does not provide a recommended CF for Ag in cephalopods, the one reported for molluscs ( $6 \times 10^4$ ) is similar to that for Zn ( $8 \times 10^4$ ) in molluscs. The IAEA CF for Zn in cephalopods is  $6 \times 10^4$  and may be considered as an approximation for Ag. Accordingly,  $^{134}\text{Cs}$ , which is also usually undetectable, was also found in the squid samples, indicating the influence of the FNA.

### 3.6. Radiological risk assessment

The European Union (EU) Erica Assessment Tool (version June 2011) was used to evaluate the radiological risk in the marine environment of the monitoring area. The Tier 1 and Tier 2 level of assessments were done. Isotopes of  $^{137}\text{Cs}$ ,  $^{134}\text{Cs}$ ,  $^{90}\text{Sr}$  and  $^{110\text{m}}\text{Ag}$  and a dose screening value of  $10 \mu\text{Gy h}^{-1}$  were selected in the assessment. The highest activity concentrations of  $^{137}\text{Cs}$ ,  $^{134}\text{Cs}$  and  $^{90}\text{Sr}$  in the seawater monitoring results were input in the assessment as the environment concentration and the parameters of distribution coefficient (Kd), concentration ratio (CR), dose conversion coefficients of radiation, occupancy factors, uncertainty factor were set as the default value in ERICA Tool. The weighting factors of internal low beta, internal beta/gamma and internal alpha were set as 3, 1 and 20.

The result of Tier 1 assessment was that "At least one value is above the  $10 \mu\text{Gy h}^{-1}$  screening dose rate. Continuing to the next tier is recommended." The output result of Tier 2 assessment is listed in Table 2, where all total dose rates per organism were lower than the screening rate by 2–3 orders of magnitude. The dose rates from  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$  were higher than those from  $^{90}\text{Sr}$  and  $^{110\text{m}}\text{Ag}$  by 2–4 orders of magnitude. It indicated that the radioactive

**Table 2**

Result of total dose rate in ERICA tool tier 2 assessment.

| Biota                                      | External dose rate – isotope [ $\mu\text{Gy h}^{-1}$ ] |         |         |         | Internal dose rate – isotope [ $\mu\text{Gy h}^{-1}$ ] |         |         |         | Total dose rate – isotope [ $\mu\text{Gy h}^{-1}$ ] |         |         |         | Total dose rate [ $\mu\text{Gy h}^{-1}$ ] per organism |
|--|--|---------|---------|---------|--|---------|---------|---------|---|---------|---------|---------|--|
|  | Ag-110m  | Cs-134  | Cs-137  | Sr-90   | Ag-110m  | Cs-134  | Cs-137  | Sr-90   | Ag-110m   | Cs-134  | Cs-137  | Sr-90   |  |
| Pelagic fish                               | 7.0E-07  | 6.1E-04 | 2.4E-04 | 8.4E-07 | 3.1E-04  | 1.2E-02 | 1.3E-02 | 4.4E-04 | 3.1E-04   | 1.3E-02 | 1.3E-02 | 4.4E-04 | 2.7E-02  |
| (Wading) bird                              | 7.0E-07  | 5.8E-04 | 2.3E-04 | 6.2E-07 | 2.9E-03  | 7.7E-02 | 7.2E-02 | 2.8E-05 | 2.9E-03   | 7.7E-02 | 7.2E-02 | 2.9E-05 | 1.5E-01  |
| Benthic fish                               | 3.8E-07  | 3.1E-04 | 1.2E-04 | 7.8E-07 | 2.6E-04  | 1.1E-02 | 1.2E-02 | 4.3E-04 | 2.6E-04   | 1.1E-02 | 1.2E-02 | 4.3E-04 | 2.4E-02  |
| Benthic mollusc                            | 3.8E-07  | 3.3E-04 | 1.3E-04 | 1.2E-06 | 1.6E-03  | 6.0E-03 | 8.2E-03 | 2.2E-03 | 1.6E-03   | 6.3E-03 | 8.3E-03 | 2.2E-03 | 1.8E-02  |
| Crustacean                                 | 3.5E-07  | 3.0E-04 | 1.2E-04 | 3.6E-07 | 1.8E-03  | 6.2E-03 | 6.1E-03 | 2.5E-04 | 1.8E-03   | 6.5E-03 | 6.2E-03 | 2.5E-04 | 1.5E-02  |
| Macroalgae                                 | 4.0E-07  | 3.4E-04 | 1.4E-04 | 3.1E-06 | 4.1E-05  | 8.6E-03 | 1.3E-02 | 5.9E-04 | 4.2E-05   | 9.0E-03 | 1.3E-02 | 5.9E-04 | 2.3E-02  |
| Mammal                                     | 3.7E-07  | 3.0E-04 | 1.2E-04 | 1.3E-07 | 9.8E-03  | 9.4E-02 | 5.7E-02 | 2.9E-05 | 9.8E-03   | 9.4E-02 | 5.7E-02 | 2.9E-05 | 1.6E-01  |
| Phytoplankton                              | 8.0E-07  | 7.5E-04 | 3.9E-04 | 2.0E-05 | 5.6E-08  | 3.3E-07 | 4.7E-07 | 5.0E-08 | 8.6E-07   | 7.5E-04 | 3.9E-04 | 2.0E-05 | 1.2E-03  |
| Polychaete worm                            | 0.0E+00  | 0.0E+00 | 0.0E+00 | 0.0E+00 | 1.2E-03  | 1.5E-02 | 2.1E-02 | 7.9E-06 | 1.2E-03   | 1.5E-02 | 2.1E-02 | 7.9E-06 | 3.7E-02  |
| Reptile                                    | 3.8E-07  | 3.1E-04 | 1.2E-04 | 1.4E-07 | 9.7E-03  | 2.0E-01 | 1.2E-01 | 2.9E-05 | 9.7E-03   | 2.0E-01 | 1.2E-01 | 2.9E-05 | 3.3E-01  |
| Sea anemones<br>or true corals –<br>colony | 3.5E-07  | 3.0E-04 | 1.2E-04 | 1.1E-06 | 4.0E-04  | 6.0E-02 | 5.7E-02 | 1.8E-02 | 4.0E-04   | 6.1E-02 | 5.7E-02 | 1.8E-02 | 1.4E-01  |
| Sea anemones<br>or true corals –<br>polyp  | 4.0E-07  | 3.4E-04 | 1.4E-04 | 2.3E-06 | 1.2E-04  | 2.9E-02 | 4.4E-02 | 1.6E-02 | 1.2E-04   | 2.9E-02 | 4.4E-02 | 1.6E-02 | 8.9E-02  |
| Vascular plant                             | 3.8E-07  | 3.3E-04 | 1.3E-04 | 1.1E-06 | 7.3E-05  | 2.2E-03 | 2.7E-03 | 7.6E-04 | 7.3E-05   | 2.5E-03 | 2.9E-03 | 7.6E-04 | 6.2E-03  |
| Zooplankton                                | 8.0E-07  | 6.9E-04 | 2.9E-04 | 1.1E-05 | 4.1E-04  | 7.0E-03 | 1.1E-02 | 4.1E-05 | 4.1E-04   | 7.7E-03 | 1.1E-02 | 5.3E-05 | 1.9E-02  |

contaminants released from the FNA would not have a significant adverse effect on marine biota at the population level, which was comparative to other research (Fisher et al., 2013; Keum et al., 2013; Kryshev et al., 2012).

#### 4. Conclusions

The monitoring results from the cruise showed that activities of  $^{137}\text{Cs}$ ,  $^{134}\text{Cs}$  and  $^{90}\text{Sr}$  were found in the surface seawater of the northwest Pacific 2 orders of magnitude higher than the background levels. The highest activity concentration of  $^{137}\text{Cs}$ , 826 mBq/L (485 times higher than the average background), was observed at the station 615 km away from Fukushima. Similar activity levels of  $^{134}\text{Cs}$  were also found in the surface seawater samples, confirming that the monitoring area was impacted by the contamination. In the biota samples of squid,  $^{134}\text{Cs}$  and  $^{110m}\text{Ag}$  were detected, which wasn't existed in the background level, indicating the radioactive release was concentrated by the marine organisms. According to the radiological assessment result of ERICA Tool, the dose rates for marine organisms in the sampling area were lower than the screening dose rate of  $10 \mu\text{Gy h}^{-1}$  by 2–3 orders of magnitude, indicating that the radioactive release from Fukushima Nuclear

Accident would not have a significant adverse effect on marine biota at the population level. However, the risk may increase while the marine biota continue concentrating the radioactive contaminants with potential transfer up the food web, especially for the organisms living in the Japanese coastal region.

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#### Appendix. Radioactivity concentration of the seawater samples

| Station | Depth (m) | Radioactivity concentration (mBq/L) |                               |                               |                  |                  |                  |                    |
|---------|-----------|-------------------------------------|-------------------------------|-------------------------------|------------------|------------------|------------------|--------------------|
|         |           | $^{137}\text{Cs}$                   | $^{134}\text{Cs}$             | $^{90}\text{Sr}$              | $^{131}\text{I}$ | $^{58}\text{Co}$ | $^{60}\text{Co}$ | $^{110m}\text{Ag}$ |
| W1-1    | 0         | $(0.29 \pm 0.02) \times 10^2$       | $(0.24 \pm 0.02) \times 10^2$ | $0.92 \pm 0.08$               | N.D.             | N.D.             | N.D.             | N.D.               |
| W1-1    | 20        | $(0.22 \pm 0.02) \times 10^2$       | $(0.18 \pm 0.01) \times 10^2$ | $1.11 \pm 0.10$               | N.D.             | N.D.             | N.D.             | N.D.               |
| W1-1    | 50        | $(0.29 \pm 0.03) \times 10^2$       | $(0.25 \pm 0.03) \times 10^2$ | $1.74 \pm 0.10$               | N.D.             | N.D.             | N.D.             | N.D.               |
| W1-2    | 0         | $(0.68 \pm 0.05) \times 10^2$       | $(0.59 \pm 0.05) \times 10^2$ | $2.84 \pm 0.10$               | N.D.             | N.D.             | N.D.             | N.D.               |
| W1-2    | 20        | $(1.33 \pm 0.11) \times 10^2$       | $(1.17 \pm 0.09) \times 10^2$ | $6.31 \pm 0.14$               | N.D.             | N.D.             | N.D.             | N.D.               |
| W1-2    | 50        | $(2.10 \pm 0.21) \times 10^2$       | $(1.88 \pm 0.19) \times 10^2$ | $6.96 \pm 0.14$               | N.D.             | N.D.             | N.D.             | N.D.               |
| W1-3    | 0         | $(1.21 \pm 0.10) \times 10^2$       | $(0.91 \pm 0.07) \times 10^2$ | $3.29 \pm 0.14$               | N.D.             | N.D.             | N.D.             | N.D.               |
| W1-3    | 20        | $(1.05 \pm 0.08) \times 10^2$       | $(0.83 \pm 0.07) \times 10^2$ | $9.87 \pm 0.18$               | N.D.             | N.D.             | N.D.             | N.D.               |
| W1-3    | 50        | $(0.70 \pm 0.06) \times 10^2$       | $(0.61 \pm 0.05) \times 10^2$ | $4.23 \pm 0.14$               | N.D.             | N.D.             | N.D.             | N.D.               |
| W1-4    | 0         | $(1.32 \pm 0.08) \times 10^2$       | $(1.17 \pm 0.07) \times 10^2$ | $1.82 \pm 0.11$               | N.D.             | N.D.             | N.D.             | N.D.               |
| W1-4    | 20        | $(0.46 \pm 0.04) \times 10^2$       | $(0.40 \pm 0.03) \times 10^2$ | $2.35 \pm 0.11$               | N.D.             | N.D.             | N.D.             | N.D.               |
| W1-4    | 50        | $(3.51 \pm 0.28) \times 10^2$       | $(3.05 \pm 0.24) \times 10^2$ | $(1.38 \pm 0.02) \times 10^1$ | N.D.             | N.D.             | N.D.             | N.D.               |
| W1-5    | 0         | $(0.53 \pm 0.04) \times 10^2$       | $(0.49 \pm 0.04) \times 10^2$ | $1.30 \pm 0.09$               | N.D.             | N.D.             | N.D.             | N.D.               |
| W1-5    | 20        | $(0.39 \pm 0.03) \times 10^2$       | $(0.33 \pm 0.03) \times 10^2$ | $8.94 \pm 0.22$               | N.D.             | N.D.             | N.D.             | N.D.               |

(continued on next page)

(continued)

| Station | Depth (m) | Radioactivity concentration (mBq/L) |                               |                               |                  |                  |                  |                           |
|---------|-----------|-------------------------------------|-------------------------------|-------------------------------|------------------|------------------|------------------|---------------------------|
|         |           | $^{137}\text{Cs}$                   | $^{134}\text{Cs}$             | $^{90}\text{Sr}$              | $^{131}\text{I}$ | $^{58}\text{Co}$ | $^{60}\text{Co}$ | $^{110\text{m}}\text{Ag}$ |
| W1-5    | 50        | $(0.52 \pm 0.04) \times 10^2$       | $(0.43 \pm 0.03) \times 10^2$ | $2.02 \pm 0.11$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| W1-6    | 0         | $(0.40 \pm 0.03) \times 10^2$       | $(0.35 \pm 0.03) \times 10^2$ | $1.62 \pm 0.11$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| W1-6    | 20        | $(0.32 \pm 0.03) \times 10^2$       | $(0.26 \pm 0.02) \times 10^2$ | $1.16 \pm 0.10$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| W1-6    | 50        | $(0.35 \pm 0.03) \times 10^2$       | $(0.30 \pm 0.02) \times 10^2$ | $3.11 \pm 0.16$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| W2-1    | 0         | $(0.25 \pm 0.02) \times 10^2$       | $(0.21 \pm 0.02) \times 10^2$ | $2.04 \pm 0.11$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| W2-1    | 20        | $(0.13 \pm 0.01) \times 10^2$       | $(0.09 \pm 0.01) \times 10^2$ | $2.10 \pm 0.10$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| W2-1    | 50        | $(0.45 \pm 0.05) \times 10^2$       | $(0.37 \pm 0.04) \times 10^2$ | $4.42 \pm 0.13$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| W2-2    | 0         | $(0.42 \pm 0.03) \times 10^2$       | $(0.22 \pm 0.02) \times 10^2$ | $1.82 \pm 0.10$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| W2-2    | 20        | $(0.15 \pm 0.01) \times 10^2$       | $(0.15 \pm 0.01) \times 10^2$ | $2.79 \pm 0.11$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| W2-2    | 50        | $(0.20 \pm 0.02) \times 10^2$       | $(0.16 \pm 0.02) \times 10^2$ | $4.24 \pm 0.15$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| W2-3    | 0         | $(1.55 \pm 0.12) \times 10^2$       | $(1.33 \pm 0.11) \times 10^2$ | $4.56 \pm 0.14$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| W2-3    | 20        | $(2.77 \pm 0.22) \times 10^2$       | $(2.30 \pm 0.18) \times 10^2$ | $(1.14 \pm 0.02) \times 10^1$ | N.D.             | N.D.             | N.D.             | N.D.                      |
| W2-3    | 50        | $(3.01 \pm 0.24) \times 10^2$       | $(2.65 \pm 0.21) \times 10^2$ | $(1.30 \pm 0.02) \times 10^1$ | N.D.             | N.D.             | N.D.             | N.D.                      |
| W2-4    | 0         | $(4.41 \pm 0.44) \times 10^2$       | $(4.02 \pm 0.40) \times 10^2$ | $(1.63 \pm 0.03) \times 10^1$ | N.D.             | N.D.             | N.D.             | N.D.                      |
| W2-4    | 20        | $(4.92 \pm 0.39) \times 10^2$       | $(4.37 \pm 0.35) \times 10^2$ | $(2.63 \pm 0.03) \times 10^1$ | N.D.             | N.D.             | N.D.             | N.D.                      |
| W2-4    | 50        | $(5.56 \pm 0.44) \times 10^2$       | $(4.94 \pm 0.40) \times 10^2$ | $(1.94 \pm 0.02) \times 10^1$ | N.D.             | N.D.             | N.D.             | N.D.                      |
| W2-5    | 0         | $(3.92 \pm 0.39) \times 10^2$       | $(3.61 \pm 0.36) \times 10^2$ | $(2.45 \pm 0.02) \times 10^1$ | N.D.             | N.D.             | N.D.             | N.D.                      |
| W2-5    | 20        | $(7.55 \pm 0.60) \times 10^2$       | $(6.61 \pm 0.53) \times 10^2$ | $9.20 \pm 0.22$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| W2-5    | 50        | $(6.78 \pm 0.54) \times 10^2$       | $(5.97 \pm 0.48) \times 10^2$ | $(2.87 \pm 0.03) \times 10^1$ | N.D.             | N.D.             | N.D.             | N.D.                      |
| W2-6    | 0         | $(1.45 \pm 0.15) \times 10^2$       | $(1.31 \pm 0.13) \times 10^2$ | $8.19 \pm 0.18$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| W2-6    | 20        | $(1.03 \pm 0.08) \times 10^2$       | $(0.90 \pm 0.07) \times 10^2$ | $6.17 \pm 0.16$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| W2-6    | 50        | $(0.80 \pm 0.06) \times 10^2$       | $(0.71 \pm 0.06) \times 10^2$ | $7.26 \pm 0.18$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| W2-7    | 0         | $(8.25 \pm 0.83) \times 10^2$       | $(7.57 \pm 0.76) \times 10^2$ | $(3.09 \pm 0.03) \times 10^1$ | N.D.             | N.D.             | N.D.             | N.D.                      |
| W2-7    | 20        | $(6.63 \pm 0.53) \times 10^2$       | $(5.83 \pm 0.47) \times 10^2$ | $(1.99 \pm 0.02) \times 10^1$ | N.D.             | N.D.             | N.D.             | N.D.                      |
| W2-7    | 50        | $(5.04 \pm 0.40) \times 10^2$       | $(4.39 \pm 0.35) \times 10^2$ | $(1.96 \pm 0.02) \times 10^1$ | N.D.             | N.D.             | N.D.             | N.D.                      |
| W3-3    | 0         | $1.37 \pm 0.13$                     | N.D.                          | $1.17 \pm 0.10$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| W3-3    | 20        | $1.99 \pm 0.16$                     | $0.65 \pm 0.05$               | $2.37 \pm 0.15$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| W3-3    | 50        | $3.33 \pm 0.33$                     | $1.76 \pm 0.18$               | $2.01 \pm 0.14$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| W3-4    | 0         | $1.76 \pm 0.17$                     | N.D.                          | $1.20 \pm 0.10$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| W3-4    | 20        | $2.05 \pm 0.16$                     | $1.08 \pm 0.09$               | $(1.26 \pm 0.02) \times 10^1$ | N.D.             | N.D.             | N.D.             | N.D.                      |
| W3-4    | 50        | $1.57 \pm 0.16$                     | $0.39 \pm 0.03$               | $1.12 \pm 0.10$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| W3-5    | 0         | $2.45 \pm 0.20$                     | $1.05 \pm 0.08$               | $1.76 \pm 0.13$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| W3-5    | 20        | $1.30 \pm 0.10$                     | N.D.                          | $1.17 \pm 0.08$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| W3-5    | 50        | $4.48 \pm 0.36$                     | $2.27 \pm 0.18$               | $2.74 \pm 0.12$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| W3-6    | 0         | $(0.35 \pm 0.04) \times 10^2$       | $(3.14 \pm 0.03) \times 10^2$ | $1.85 \pm 0.09$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| W3-6    | 20        | $4.14 \pm 0.33$                     | $1.93 \pm 0.15$               | $1.82 \pm 0.10$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| W3-6    | 50        | $(1.50 \pm 0.12) \times 10^2$       | $(1.31 \pm 0.10) \times 10^2$ | $1.95 \pm 0.11$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| W3-7    | 0         | $(1.94 \pm 0.19) \times 10^2$       | $(1.75 \pm 0.17) \times 10^2$ | $9.69 \pm 0.15$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| W3-7    | 20        | $(2.58 \pm 0.21) \times 10^2$       | $(2.26 \pm 0.18) \times 10^2$ | $8.43 \pm 0.26$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| W3-7    | 50        | $(4.85 \pm 0.39) \times 10^2$       | $(4.36 \pm 0.35) \times 10^2$ | $(1.80 \pm 0.02) \times 10^1$ | N.D.             | N.D.             | N.D.             | N.D.                      |
| W3-8    | 0         | $(1.76 \pm 0.18) \times 10^2$       | $(1.59 \pm 0.16) \times 10^2$ | $8.55 \pm 0.16$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| W3-8    | 20        | $(1.89 \pm 0.15) \times 10^2$       | $(1.68 \pm 0.13) \times 10^2$ | $7.85 \pm 0.18$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| W3-8    | 50        | $(1.99 \pm 0.16) \times 10^2$       | $(1.69 \pm 0.13) \times 10^2$ | $8.72 \pm 0.17$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| W3-9    | 0         | $(2.82 \pm 0.28) \times 10^2$       | $(2.57 \pm 0.26) \times 10^2$ | $8.74 \pm 0.17$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| W3-9    | 20        | $(1.68 \pm 0.13) \times 10^2$       | $(1.44 \pm 0.12) \times 10^2$ | $9.19 \pm 0.18$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| W3-9    | 50        | $(2.84 \pm 0.23) \times 10^2$       | $(2.55 \pm 0.20) \times 10^2$ | $(1.03 \pm 0.02) \times 10^1$ | N.D.             | N.D.             | N.D.             | N.D.                      |
| Wa      | 0         | $(4.64 \pm 0.37) \times 10^2$       | $(4.09 \pm 0.33) \times 10^2$ | $4.74 \pm 0.14$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| Wa      | 20        | $(0.47 \pm 0.05) \times 10^2$       | $(0.39 \pm 0.04) \times 10^2$ | $1.25 \pm 0.10$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| Wa      | 50        | $(0.18 \pm 0.01) \times 10^2$       | $(0.15 \pm 0.01) \times 10^2$ | $2.34 \pm 0.12$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| Wb      | 0         | $(3.22 \pm 0.26) \times 10^2$       | $(2.80 \pm 0.22) \times 10^2$ | $1.76 \pm 0.14$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| Wb      | 20        | $(0.22 \pm 0.02) \times 10^2$       | $(0.19 \pm 0.01) \times 10^2$ | $1.25 \pm 0.11$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| Wb      | 50        | $(0.23 \pm 0.02) \times 10^2$       | $(0.19 \pm 0.02) \times 10^2$ | $2.81 \pm 0.13$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| J4-1    | 0         | $1.63 \pm 0.13$                     | $1.18 \pm 0.09$               | $2.33 \pm 0.11$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| J4-1    | 20        | $2.58 \pm 0.21$                     | $2.44 \pm 0.20$               | $1.63 \pm 0.10$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| J4-1    | 50        | $2.16 \pm 0.22$                     | $0.78 \pm 0.08$               | $1.46 \pm 0.12$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| J4-2    | 0         | $2.63 \pm 0.21$                     | $0.77 \pm 0.06$               | $2.40 \pm 0.13$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| J4-2    | 20        | $5.69 \pm 0.46$                     | $4.04 \pm 0.32$               | $2.08 \pm 0.12$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| J4-2    | 50        | $2.75 \pm 0.27$                     | $0.98 \pm 0.09$               | $1.65 \pm 0.10$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| J4-3    | 0         | $2.12 \pm 0.17$                     | $1.51 \pm 0.12$               | $2.12 \pm 0.11$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| J4-3    | 20        | $3.32 \pm 0.27$                     | $1.71 \pm 0.14$               | $1.41 \pm 0.12$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| J4-3    | 50        | $3.53 \pm 0.35$                     | $1.57 \pm 0.16$               | $1.50 \pm 0.12$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| J4-4    | 0         | $2.30 \pm 0.18$                     | $1.07 \pm 0.09$               | $3.58 \pm 0.13$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| J4-4    | 20        | $4.41 \pm 0.35$                     | $3.05 \pm 0.24$               | $1.27 \pm 0.09$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| J4-4    | 50        | $4.90 \pm 0.49$                     | $2.75 \pm 0.27$               | $1.36 \pm 0.14$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| J4-5    | 0         | $1.92 \pm 0.15$                     | $1.78 \pm 0.14$               | $1.93 \pm 0.12$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| J4-5    | 20        | $1.58 \pm 0.13$                     | $0.61 \pm 0.05$               | $2.15 \pm 0.12$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| J4-5    | 50        | $2.75 \pm 0.27$                     | N.D.                          | $1.36 \pm 0.14$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| J4-6    | 0         | $0.62 \pm 0.05$                     | $0.22 \pm 0.02$               | $2.19 \pm 0.11$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| J4-6    | 20        | $1.57 \pm 0.16$                     | N.D.                          | $1.66 \pm 0.14$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| J4-6    | 50        | $2.49 \pm 0.20$                     | $0.46 \pm 0.04$               | $1.55 \pm 0.12$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| J4-7    | 0         | $(0.30 \pm 0.02) \times 10^2$       | $(0.23 \pm 0.02) \times 10^2$ | $2.62 \pm 0.11$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| J4-7    | 20        | $1.96 \pm 0.20$                     | $0.98 \pm 0.09$               | $2.08 \pm 0.12$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| J4-7    | 50        | $4.20 \pm 0.34$                     | $1.63 \pm 0.13$               | $1.87 \pm 0.15$               | N.D.             | N.D.             | N.D.             | N.D.                      |
| J4-8    | 0         | $(2.07 \pm 0.17) \times 10^2$       | $(1.79 \pm 0.14) \times 10^2$ | $9.85 \pm 0.18$               | N.D.             | N.D.             | N.D.             | N.D.                      |

(continued)

| Station                                     | Depth (m) | Radioactivity concentration (mBq/L) |                               |                                 |                  |                  |                  |                    |
|---|-----------|-------------------------------------|-------------------------------|---------------------------------|------------------|------------------|------------------|--------------------|
|   |           | <sup>137</sup> Cs                   | <sup>134</sup> Cs             | <sup>90</sup> Sr                | <sup>131</sup> I | <sup>58</sup> Co | <sup>60</sup> Co | <sup>110m</sup> Ag |
| J4-8  | 20        | $(2.04 \pm 0.20) \times 10^2$       | $(1.85 \pm 0.19) \times 10^2$ | $8.09 \pm 0.15$                 | N.D.             | N.D.             | N.D.             | N.D.               |
| J4-8  | 50        | $(3.94 \pm 0.32) \times 10^2$       | $(3.54 \pm 0.28) \times 10^2$ | $(1.18 \pm 0.02) \times 10^1$   | N.D.             | N.D.             | N.D.             | N.D.               |
| J4-9  | 0         | $(0.46 \pm 0.04) \times 10^2$       | $(0.39 \pm 0.03) \times 10^2$ | $2.22 \pm 0.10$                 | N.D.             | N.D.             | N.D.             | N.D.               |
| J4-9  | 20        | $(0.37 \pm 0.04) \times 10^2$       | $(0.35 \pm 0.04) \times 10^2$ | $3.09 \pm 0.10$                 | N.D.             | N.D.             | N.D.             | N.D.               |
| J4-9  | 50        | $(2.59 \pm 0.21) \times 10^2$       | $(2.33 \pm 0.19) \times 10^2$ | $2.24 \pm 0.10$                 | N.D.             | N.D.             | N.D.             | N.D.               |
| J4-10                                       | 0         | $(0.51 \pm 0.04) \times 10^2$       | $(0.45 \pm 0.04) \times 10^2$ | $1.69 \pm 0.10$                 | N.D.             | N.D.             | N.D.             | N.D.               |
| J4-10                                       | 20        | $(0.51 \pm 0.05) \times 10^2$       | $(0.45 \pm 0.05) \times 10^2$ | $1.04 \pm 0.12$                 | N.D.             | N.D.             | N.D.             | N.D.               |
| J4-10                                       | 50        | $(0.70 \pm 0.06) \times 10^2$       | $(0.59 \pm 0.05) \times 10^2$ | $3.34 \pm 0.17$                 | N.D.             | N.D.             | N.D.             | N.D.               |
| J4-11                                       | 0         | $(0.53 \pm 0.04) \times 10^2$       | $(0.45 \pm 0.04) \times 10^2$ | $2.51 \pm 0.11$                 | N.D.             | N.D.             | N.D.             | N.D.               |
| J4-11                                       | 20        | $(0.51 \pm 0.05) \times 10^2$       | $(0.45 \pm 0.05) \times 10^2$ | $2.20 \pm 0.14$                 | N.D.             | N.D.             | N.D.             | N.D.               |
| J4-11                                       | 50        | $(0.52 \pm 0.05) \times 10^2$       | $(0.43 \pm 0.03) \times 10^2$ | $1.67 \pm 0.10$                 | N.D.             | N.D.             | N.D.             | N.D.               |
| J4-12                                       | 0         | $(0.49 \pm 0.04) \times 10^2$       | $(0.42 \pm 0.03) \times 10^2$ | $5.90 \pm 0.19$                 | N.D.             | N.D.             | N.D.             | N.D.               |
| J4-12                                       | 20        | $(0.47 \pm 0.05) \times 10^2$       | $(0.39 \pm 0.04) \times 10^2$ | $1.54 \pm 0.11$                 | N.D.             | N.D.             | N.D.             | N.D.               |
| J4-12                                       | 50        | $(0.60 \pm 0.05) \times 10^2$       | $(0.51 \pm 0.04) \times 10^2$ | $1.94 \pm 0.11$                 | N.D.             | N.D.             | N.D.             | N.D.               |
| J4-13                                       | 0         | $(1.14 \pm 0.09) \times 10^2$       | $(0.98 \pm 0.07) \times 10^2$ | $6.92 \pm 0.18$                 | N.D.             | N.D.             | N.D.             | N.D.               |
| J4-13                                       | 20        | $(0.43 \pm 0.04) \times 10^2$       | $(0.39 \pm 0.04) \times 10^2$ | $2.09 \pm 0.13$                 | N.D.             | N.D.             | N.D.             | N.D.               |
| J4-13                                       | 50        | $(0.60 \pm 0.05) \times 10^2$       | $(0.52 \pm 0.04) \times 10^2$ | $1.80 \pm 0.14$                 | N.D.             | N.D.             | N.D.             | N.D.               |
| Detection limit                             |           | 0.26                                | 0.23                          | 0.24                            |                  |                  |                  |                    |
| Coastal background <sup>a</sup> (2000–2010) |           | 0.04–3.4<br>1.7 ± 0.6 (n = 961)     | N.D.                          | 0.01–2.6<br>1.2 ± 0.4 (n = 871) | N.D.             | N.D.             | N.D.             | N.D.               |

N.D. — activity was lower than LLD.

<sup>a</sup> From IAEA MARIS Database.

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